



Seasonal Variation in Heavy Metal Content of Tongue Sole, *Cynoglossus brownii* and Croaker, *Pseudotolithus typus* from Lagos and Delta States, Nigeria

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Authors' contributions

This project was conceived by Author FG who suggested the constitution of a research team headed by author AT and comprising authors FG and OD for evaluation of quality and safety of seafood in Nigeria. Author OR collected experimental samples and carried out the laboratory analyses with the assistance of other authors. Statistical analyses were carried out by author FG, while the first manuscript was drafted by author OR. All authors approved the contents of the final paper for publication in BJAST.

Research Article

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ABSTRACT

Aims: The aims of this study were to determine the levels of lead (Pb), Cadmium (Cd), Copper (Cu), Chromium (Cr) and Zinc (Zn) in two economically important Nigerian fish species, tongue sole, *Cynoglossus brownii* and Croaker, *Pseudotolithus typus* from Makoko and Koko in Lagos and Delta states respectively; compare the levels of these metals with recommended international tolerable levels thereby creating awareness on potential health risks of human consumption of these fish species.

Study Design: The study locations were purposively selected, being coastal locations where major industries and crude oil exploration facilities are located; and with a high probability of discharging effluents with high levels of heavy metals. Twelve samples each

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of both fish species were purchased from landing sites in Makoko (Lagos State) and Koko (Delta State) respectively and transported in ice boxes to the laboratories for subsequent analyses.

Place and Duration of Study: Department of Environmental Management and Toxicology, Federal University of Agriculture, Abeokuta (FUNAAB) between May, 2010 and April, 2011.

Methodology: Twelve samples each of tongue sole and croaker were collected bi-monthly from each sampling location in May, July and September, 2010 (Wet season) and November (2010) and January and March, 2011 (Dry season). Six samples each were analyzed for heavy metal contents using Atomic Absorption Spectrophotometry (AAS) after wet digestion of dried, milled samples of the fish muscles with 1:1 HNO₃: H₂O₂.

Results: Metal contents (µg/g, dry weight) for tongue sole in Lagos and Delta states during the wet season (April – October) were: Pb (0.330 and 0.149), Cd (0.097 and 0.063), Cu (0.869 and 0.434), Cr (1.184 and 0.826), Zn (7.612 and 7.890); for croaker: Pb (0.163 and 0.045), Cd (0.115 and 0.029), Cu (0.990 and 0.694), Cr (1.008 and 1.038) and Zn (6.756 and 6.168). For the dry season (November – March) metal contents obtained were: Pb (0.534 and 0.527), Cd (0.373 and 0.410), Cu (0.437 and 0.556), Cr (0.834 and 0.983), Zn (6.764 and 9.343) for sole; and Pb (0.210 and 0.213), Cd (0.264 and 0.307), Cu (0.442 and 0.483), Cr (0.577 and 0.597) and Zn (4.783 and 5.924) for croaker. Significant ($P = 0.05$) variations were observed in metal content between sole and croaker and also between dry and wet seasons notably for Cd, Cr and Cu. Generally, metal concentrations were higher in sole than croaker; and Cadmium content was significantly ($P = 0.05$) higher in fish samples during wet season, while Chromium and Copper were significantly ($P = 0.05$) higher during the dry season. Cd and Cr levels in the test species were found to exceed recommended international tolerable levels.

Conclusion: Heavy metal contents varied among the fish species evaluated and also between seasons. During the dry season, chromium and copper contents of fish were significantly higher, while Cadmium content of fish was significantly higher during the wet season. Cadmium and Chromium contents exceeded international tolerable limits, thus continued consumption of these fish species could expose consumers to serious health risks arising from metal toxicity. A programme of continuous monitoring and control of the physical and chemical characteristics of the components and food products of Nigeria's coastal zone is recommended as a management tool for this important ecological zone.

Keywords: Heavy metal; *Cynoglossus brownii*; *Pseudotolithus typus*; Lagos; Delta; dry season; wet season; health risk.

1. INTRODUCTION

Tongue sole, *Cynoglossus brownii* and croaker, *Pseudotolithus typus* are among the most commonly available and economically important fish species in Nigeria's maritime states. They contribute significantly to the animal protein intake of the urban population in these states. Compared to other coastal states, Lagos and Delta states are peculiar, with a blossoming shipping and freighting industry especially Lagos state, which doubles as Nigeria's industrial and commercial centre, having the highest number of industries and arguably the largest human population within a state in the country. As the commercial and industrial nerve centre of Nigeria, with an estimated population of over 9 million people [1,2], environmental concerns are normally focused on Lagos State. Over 60% of Nigeria's industries are cited in the state, each discharging its characteristic range of effluents, often containing heavy metals into the state's terrestrial and aquatic ecosystems. Water pollutants generally originate from definite or point; and dispersed (non point) sources. While point

source pollutants are distinct and confined, including accidental spills from industrial sites that empty into streams or rivers, non-point sources, such as run-off, are diffuse and intermittent; and largely influenced by factors such as land use, climate, hydrology, topography, native vegetation and geology [3,4]. Common urban non point sources of pollution include urban run-off from streets or agricultural fields, characteristically containing a variety of pollutants like heavy metals, chemicals and sediments. Rural sources of non point pollutants are generally associated with agriculture, mining or forestry. Non point sources are difficult to monitor and control [5]. The hazard presented by a particular water pollutant depends on several factors including the concentration or toxicity of the pollutant in the environment, and the degree of exposure to people or other organisms [6]. It is estimated that over 80% of coastal pollution originates from land-based sources, including industrial, agricultural, oil and gas production and marine oil transportation activities [7]. Being a universal solvent, water would naturally dissolve toxic materials dumped into it, thereby putting living organisms in the water at risk. Recent works suggest that toxic materials threaten the ocean bottom as well as the entire marine ecosystem. The base of the marine food chain consists of planktonic life which occupies the upper 3mm of the ocean water. The young of most fin and shell fishes also reside in the upper few millimeters of the ocean; and tend to concentrate pollutants such as toxic chemicals and heavy metals. It is reported that concentrations of zinc, lead, and copper in the upper 3mm (or micro layer) is from 10 to 1000 times higher than in the deepest waters due to a process referred to as surface film enrichment [8]. Thus there is fear that disproportionate pollution of this micro layer will have serious effects on marine organisms [9,10,11], resulting in major impacts on people and society as contaminated marine organisms transmit toxic elements or diseases to their consumers [9].

Heavy metal contamination of the aquatic environment has become a global challenge, mainly because heavy metals are not degradable; and most of them could have toxic effects not only on aquatic flora but also on consumers of aquatic food products [12]; they are therefore of particular concern in view of their persistence, potential toxic effects, indestructibility and ability to bio-accumulate in aquatic ecosystems [13,14]). Heavy metal concentrations in aquatic ecosystems are usually monitored by measuring their concentrations in water, sediments and biota [15,16]. Generally metal concentrations are low in water and attain considerably higher concentrations in sediments and biota [17]. Concerns over heavy metal pollution of the marine environment have been rising due to the human activities that contribute significantly to the release of these metals into the environment. It is reported [18] that heavy metals in the environment are brought about primarily by anthropogenic sources. Though heavy metals are a part of the natural environment and provide numerous benefits to society, those that get deposited in water bodies result from coal and oil combustion, internal combustion engines, local point sources, and direct deposition from air pollution, bedrock geology and soils [18].

The concern posed by heavy metals in the aquatic environment creates an immense threat to the existence of organisms thriving in the area and the ecological integrity of the habitat, particularly as heavy metals may enter the food chains, persist in the environment, bio-accumulate and bio-magnify, then increase exposure of consumers of aquatic products to serious health risks.

Croaker, *Pseudotolithus typus* and sole, *Cynoglossus brownii* are major components of fish landings from Nigerian coastal states, among which Lagos and Delta states are identified as being highly productive [19,20,21]. Apart from their relative, abundance in the Nigerian artisanal fisheries, both sole and croaker are relished by consumers locally and

internationally; and sole fillets feature prominently as export products to Europe. Generally, the two experimental fish species have high economic and nutritional significance in Nigeria.

The present study was carried out to evaluate seasonal variability in level of some common heavy metal pollutants (cadmium, chromium, lead, copper and zinc) in the flesh of croaker and sole; compare the levels of these metals in the fish species with international tolerable levels and finally provide information on the quality of the fish species; and possible health challenges inherent in the consumption of fish with high levels of heavy metals.

2. MATERIALS AND METHODS

Two fish species, *Pseudotolithus typus* (croaker) and *Cynoglossus brownii* (sole) which are lentic and benthic fishes respectively were used for the study. Fish samples were procured from fishermen at landing sites at Makoko, located in the Lagos Mainland Local Government Area, LGA, Lagos State ($6^{\circ} 29' 44''$ N $3^{\circ} 23' 39''$ E); and Koko ($6^{\circ} 0' 11.31''$ N $5^{\circ} 28' 32.36''$ E) in the Warri North LGA, Delta State, Nigeria as shown in Fig. 1.

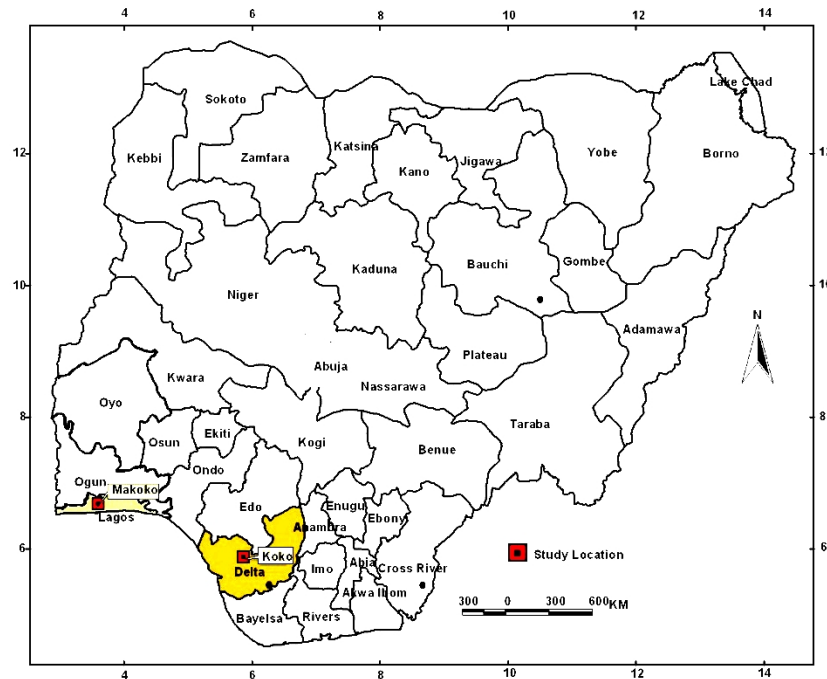


Fig. 1. Map showing sampling locations, Makoko, Lagos and Koko, Delta states

Sampling was done six times in May, July, September (Wet season) and November, January, and March (Dry season) at Makoko, Lagos State and Koko, Delta State, Nigeria. Fish samples were procured fresh from fishermen at landing sites in the study locations; and preserved by cooling in ice buckets till they were transferred into deep freezers in the laboratory and kept frozen at -20°C until analysis.

Prior to analysis frozen fish samples were allowed to defrost at room temperatures for about two hours. The thawed fish samples were oven dried at 105°C for about 12h until a constant

weight was obtained. Thereafter dried fish samples were homogenized using a ceramic mortar and pestle. Samples were prepared with utmost care to avoid contamination. Hand-gloves were worn throughout the analytical procedures, and precautions were taken to carry out all preparations under strict sanitary laboratory conditions. Where drying was not completed in one day, samples were kept in desiccators and oven-drying was continued the following day. All glass wares used were washed and rinsed severally with distilled water; then washed in 10% HCl before use.

Samples were digested according to the wet digestion procedure [22], by digesting 10.00 g of dried sample in 60 ml of freshly prepared 1:1 HNO₃/ H₂O₂ solution at 160 °C on a hot plate for about one hour until the volume of contents was reduced to about 5 ml. Contents were then filtered and the filtrate transferred to a standard flask and made up to 25 ml with distilled de-ionized water. This was stored in plastic bottles and the concentrations of Pb, Cr, Cu, Cd and Zn were evaluated using air-acetylene flame AAS (AAnalyst 200, Perkin Elmer).

Data obtained were subjected to one-way Analysis of Variance (ANOVA) [23]; and significantly different means were separated using the methods of Duncan. The values obtained were presented as Least Significance Differences (LSD) of means at $P = 0.05$.

3. RESULTS AND DISCUSSION

Levels of Pb, Cd, Cu, Cr and Zn observed in the Sole and Croaker during the dry season (November – March) at both study locations compared with international tolerable values are presented in Table 1; while those for the wet season (April – October) are presented in Table 2.

Table 1. Heavy metal content of fish samples during the dry season (November–March)

Metals	Metal content (µg/g) of fish samples*				International standards
	LS	LC	DS	DC	
Pb	0.330±0.31	0.163±0.05	0.149±0.13	0.045±0.05	0.200 [24]
Cd	0.097±0.03	0.115±0.09	0.063±0.05	0.029±0.03	0.050 [24]
Cu	0.869±0.48	0.990±0.57	0.434±0.22	0.694±0.29	3.280 [25]
Cr	1.184±0.60	1.008±0.15	0.826±0.18	1.038±0.12	0.730 [25]
Zn	7.612±0.83	6.756±0.83	7.890±0.80	6.168±0.34	30.000 [24]

*LS (Lagos sole), LC (Lagos Croaker), DS (Delta sole), DC (Delta croaker)

Table 2. Heavy metal content of fish samples during the wet season (April–October)

Metals	Metal content (µg/g) of fish samples*				International standards
	LS	LC	DS	DC	
Pb	0.534±0.62	0.210±0.14	0.527±0.61	0.213±0.27	0.200 [24]
Cd	0.373±0.14	0.264±0.15	0.416±0.24	0.307±0.18	0.050 [24]
Cu	0.437±0.14	0.442±0.23	0.556±0.15	0.483±0.18	3.280 [25]
Cr	0.834±0.32	0.517±0.37	0.983±0.36	0.597±0.44	0.730 [25]
Zn	6.764±1.56ab	4.783±1.87b	9.343±2.82a	5.724±2.08ab	30.000 [24]

*LS (Lagos sole), LC (Lagos Croaker), DS (Delta sole), DC (Delta croaker). Rows with different superscripts are significantly ($P < 0.05$) different.

Pb ranged from 0.045 (DC) to 0.330 $\mu\text{g/g}$ (LS) during the dry season; with the level in LS being higher than tolerable International standard (0.200 $\mu\text{g/g}$) during the season. Cd levels were higher in all experimental samples except DC during the dry season; and ranged from 0.029 (DC) to 0.115 $\mu\text{g/g}$ (LC). Cu ranged from 0.434 (DS) to 0.990 $\mu\text{g/g}$ (LC) but was lower in all experimental samples than the International tolerable value of 3.280 $\mu\text{g/g}$ [25]. Cr levels were higher in all experimental fish samples during the dry season than the International tolerable value of 0.730 $\mu\text{g/g}$ [25], ranging from 0.826 $\mu\text{g/g}$ in DS to 1.184 in LS. Zn level ranged from 6.168 $\mu\text{g/g}$ in DC to 7.612 $\mu\text{g/g}$ in LS; but were below the International tolerable value of 30.000 [24].

Heavy metal content of experimental fish species was not significantly different between species or location during the dry season. As shown on table 2, levels of Pb (0.210 – 0.534 $\mu\text{g/g}$ in LC and LS respectively) and Cd (0.264 – 0.416 $\mu\text{g/g}$ in LC and DS respectively) were higher in experimental fish during wet season than tolerable levels of 0.200 (Pb) and 0.050 (Cd) according to [24]. Also, levels of Pb and Cd in sole during the dry season were similar in both study locations; but higher than tolerable values, while Cr levels were higher in Delta than Lagos state. Levels of Cu and Zn in sole were higher in Delta during the dry season, but lower than tolerable values. Similar trends as reported above were observed for all trace metals in croaker during the dry season. With the exception of Cu and Zn, trace metal content of sole was higher in Lagos than Delta state; with Pb and Cr levels being higher than reported tolerable levels during the wet season. Zn and Cu levels of Croaker were higher in Lagos than Delta state, but lower than tolerable values during the wet season.

Table 3 describes variation in metal contents between wet and dry seasons. Generally, significant ($P = 0.5$) variation was observed between these seasons in Cd, Cu and Cr levels, with Cu and Cr being higher during the dry than wet season; and Cd being higher during the wet than the dry season.

Table 3. Seasonal variation in metal content of sole and croaker

Season	*Metal contents ($\mu\text{g/g}$)				
	Pb	Cd	Cu	Cr	Zn
Wet	0.34 \pm 0.11	0.34 \pm 0.04 a	0.48 \pm 0.04 b	0.73 \pm 0.10 b	6.70 \pm 0.64
Dry	0.17 \pm 0.05	0.08 \pm 0.02 b	0.75 \pm 0.12 a	1.01 \pm 0.09 a	7.11 \pm 0.30

*Columns with different superscripts are significantly ($P < 0.05$) different

Variability of metal content of fish with season was established in the present study in line with previous studies [26,27,28]. While Cadmium content was higher during the wet season, copper and chromium levels were higher during the dry season; and zinc and lead contents were similar at both seasons.

Table 4 describes variation in metal content between experimental fish species. Pb and Zn were significantly ($P = 0.5$) higher in Sole than in Croaker.

Table 4. Variation in metal content between fish species

Fish species	*Metal contents ($\mu\text{g/g}$)				
	Pb	Cd	Cu	Cr	Zn
Sole	0.41 \pm 0.12 a	0.26 \pm 0.06	0.56 \pm 0.08	0.95 \pm 0.10	7.92 \pm 0.51 a
Croaker	0.13 \pm 0.04 b	0.19 \pm 0.04	0.62 \pm 0.10	0.76 \pm 0.10	5.83 \pm 0.44 b

*Columns with different superscripts are significantly ($P < 0.05$) different

As shown below (Table 5), observed differences in metal content of Sole and Croaker between Lagos and Delta states were not statistically significant.

Table 5. Variation in metal content of Sole and Croaker between Lagos and Delta states

Location (states)	Metal contents ($\mu\text{g/g}$)				
	Pb	Cd	Cu	Cr	Zn
Delta	0.28±0.10	0.23±0.04	0.65±0.11	0.86±0.11	6.38±0.46
Lagos	0.27±0.11	0.23±0.06	0.54±0.05	0.85±0.09	7.37±0.61

Metal (Pb, Cd, Cu, Cr and Zn) concentrations in the two fish species and sampling locations were compared with international standards. As expected, the metals were more concentrated in sole, a benthic fish than croaker, a pelagic fish; except for Cu where the concentrations in croaker were higher than in sole; and Cd where concentrations found in Croaker from Lagos State (LC) was higher than those found in Sole. The higher concentrations of heavy metals in sole, a benthic fish confirmed reports that metals concentrate more in the sediments and benthic region of aquatic environments [17,29] rather than the water column. Species variation in metal accumulation had also been widely reported [30,31,32], thus the observed affinity for croaker to accumulate Cu and Cd in higher quantities than sole.

In general, lead values obtained in this study were similar to those reported in previous studies on freshwater fish species in Nigeria [33], but lower than those reported for fresh and processed *Tilapia* and *Chrysichthys* species in Ghana [34]; and higher than levels in various freshwater fish species in the Philippines [31]. Lead levels in both experimental fish and study locations exceeded international tolerable levels during the wet season, but only in Lagos sole in the dry season, possibly due to increased influx of metal pollutants from run-off into the aquatic environment during the wet season. Lead is known to exert its most significant effects on the nervous system, the hematopoietic system and the kidney. It has effects on the nervous system including motor disturbances.

The concentration of Cadmium in all the experimental fish were higher than the international tolerable level of $0.05\mu\text{g/g}$ [24], except in Delta croaker during the dry season with a concentration of $0.029\mu\text{g/g}$. Cadmium concentrations in this study were higher than those earlier reported [28,29] and exceeded the international tolerable limit as noted in the Philippine study [30]. Adverse effects of cadmium exposure may include chronic renal failure, kidney stones, liver damage, lung cancer, osteomalacia, increased possibility of hypertension, prostate cancer, and proteinuria. Chronic cadmium exposure has been reported to cause mild anemia, skeletal lesions, Itai-itai disease, anosmia, and yellowing of teeth [30].

Chromium concentrations in the present study were found to be lower than levels previously reported [35], but higher than the IAEA standard of $0.730\mu\text{g/g}$ for chromium [25]; and those reported [28] for some Nigerian freshwater fish species including *Clarias gariepinus*, *C. anguillaris* and *Oreochromis niloticus*. Reported health effects of chromium toxicity include haemolysis, renal and liver failure, cancer and damage to circulatory and nervous tissues [35,36,37].

Mean concentrations of zinc in this study were lower than the reported standard of 3.280ppm [25]. Zinc deficiency has been associated with dermatitis, anorexia, growth retardation; poor wound healing, hypogonadism with impaired reproductive capacity.

Also, the study showed more heavy metal pollution in fish samples from Lagos than those from Koko, except for Zn, where the concentrations in Delta were more than those of Lagos state. Zinc concentrations in experimental samples were much lower than the international tolerable limits.

4. CONCLUSION

Trace metal contents of the two economically important fish species evaluated varied with season and among species, with the benthic species studied (Sole) having significant higher levels of Pb and Zn than the pelagic species, Croaker. Cu and Cr levels were significantly higher during the dry season; and Cd level in fish species were significantly higher during the wet season. Reported studies on variation in metal content of economically important Nigerian fish species are generally lacking especially for marine species; thus it is important that findings of this research be published to educate consumers on possible health hazards associated with fish consumption.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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